# Light Induced Inverse-Square Law Interactions between Nanoparticles: "Mock Gravity" at the Nanoscale

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The interaction forces between identical resonant molecules or nanoparticles, optically induced by a quasimonochromatic isotropic random light field, are theoretically analyzed. In general, the interaction force exhibits a far-field oscillatory behavior at separation distances larger than the light wavelength. However, we show that the oscillations disappear when the frequency of the random field is tuned to an absorption Fröhlich resonance, at which the real part of the particle's electric polarizability is zero. At the resonant condition, the interaction forces follow a long-range gravitylike inverse square distance law which holds for both near- and far-field separation distances.

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*Introduction.*—The origin of long-range attractive interactions has fascinated scientists for centuries. The remarkable Fatio de Duillier–LeSage [1,2] corpuscular theory, introduced as early as 1690 and generalized to electromagnetic waves by Lorentz [3], proposed that, due to their mutual shadowing, two absorbing particles in an isotropic radiation field experience an attractive force that follows a gravitylike inverse square distance law. Similar "Mock gravity" interactions were later introduced by Spitzer [4] and Gamow [5] in the context of Galaxy formation but their actual relevance in cosmology has never been unambiguously established [6,7]. Recent work [8–10] demonstrated that the interaction force between nonabsorbing dielectric particles and atoms in a quasimonochromatic isotropic random light field always presents an oscillatory behavior for distances larger than the light wavelength. Our main goal here is to show that, under specific resonant conditions, these forces become nonoscillating, $1/r^2$, attractive forces.

The interaction between two objects is usually defined to be long ranged if the force decays with their distance apart, $r$, as a power law $\sim 1/r^{n+1}$ with $n$ smaller than the spatial dimension of the system. Gravity is a typical example of a long-range attractive force in three dimensions while the interaction between electric or magnetic dipoles ($n = 3$) is borderline in between short and long range attraction [11]. In contrast, dispersion forces between nonpolar, neutral molecules and particles are short range: at close distances the Coulomb interaction between the fluctuating electric dipole moments leads to the van deer Waals–London interaction energy [12] proportional to $1/r^6$. However, when $r$ is larger than a characteristic resonance wavelength $\lambda_F$, retardation effects become important since the dipole moments fluctuate many times over the period the light takes to pass between particles. The interaction energy varies then as $1/r^7$ as first shown by Casimir and Polder [13]. These interactions can also be derived as a special case of Lifshitz's theory of attraction between macroscopic bodies [14] induced by *equilibrium* quantum and thermal electromagnetic field fluctuations [14–17].

In the last years there has been an increasing interest in understanding the nonequilibrium analogs of Casimir forces arising in the interaction between bodies at different temperature [18,19] like those induced by blackbody radiation from a hot source on atoms and nanoparticles [20–22]. Surprisingly strong long-range interactions between atoms or nonabsorbing dielectric particles in a quasimonochromatic fluctuating random field were predicted [8–10] and experimentally demonstrated for micron-sized particles [10] (similar interactions between pairs of dipoles under the excitation of multiple laser beams were also discussed [23]). Although the effective interaction range can be controlled by the spectral bandwidth of the fluctuating field [10,24,25] (with the Casimir-Lifshsitz interaction recovered in the limit of a quantum black body spectrum [10]), the existence of three-dimensional artificial gravitylike, inverse square law, interaction forces had not yet been demonstrated.

For small separation distances, gravitylike interactions were first predicted by quantum electrodynamics (QED) calculations for atoms and molecules [8,23] and later by a classical approach on Rayleigh nanoparticles [9] leading to analogous results. In these studies, the imaginary part of the polarizability was either not included in the calculations [8,23] or was taken into account but considered negligible [9]; i.e., radiation pressure effects were neglected. However, as discussed below, these effects dominate the near-field interactions of nonabsorbing particles, leading to a different interaction law.

In this Letter we show that, in contrast with atoms or dielectric particles, the interaction force between two identical resonant molecules or plasmonic nanoparticles, whose extinction cross section is dominated by absorption, can follow a true attractive inverse square law all the way from near to far-field separation distances. In his celebrated "Lectures on Theoretical Physics" [3], Lorentz already suggested that electrodynamic interactions in the presence of absorption could lead to an inverse square law interaction force similar to the original Fatio-Lesage's corpuscular theory. However, as we will see, the ideal nonoscillating $\sim 1/r^2$ law can only be achieved when the frequency of the random field is tuned to the particles' Fröhlich resonance [26] (e.g., the Fröhlich frequency, $\omega_F$, of plasmonic silver nanoparticles).

*Optical response of the nanoparticles: polarizability.*—To this end, let us consider two identical molecules or nanoparticles in an otherwise homogeneous medium with refractive index $n_h = 1$. Following a standard classical approach, based on the macroscopic Maxwell equations, we assume that their optical response, at a frequency $\omega$, is characterized by a scalar electrical polarizability $\alpha(\omega)$,

$$\alpha(\omega) = \left(\alpha_0^{-1}(\omega) - i\frac{k^3}{6\pi}\right)^{-1} = |\alpha(\omega)|e^{i\delta_\omega} \tag{1}$$

[where $\delta_\omega$ the scattering phase-shift, $k = \omega/\mathrm{c}$ (with c being the speed of light in vacuum)], with extinction cross section $\sigma_{\text{ext}} = k\text{Im}\{\alpha\}$ and absorption cross section

$$\sigma_{\text{abs}} = k\left\{\text{Im}\{\alpha\} - \frac{k^3}{6\pi}|\alpha|^2\right\} = k|\alpha|^2\frac{\text{Im}\{\alpha_0\}}{|\alpha_0|^2} \geq 0 \tag{2}$$

[for nanospheres, $\alpha$ is proportional to the first Mie electric coefficient $a_1$ [26], $\alpha \equiv ia_1(6\pi/k^3)$]. $\alpha_0(\omega)$ is the quasistatic polarizability which, if the particle's radius $a$ is sufficiently small, $ka \ll 1$, is given by $\alpha_0(\omega) = 4\pi a^3[\epsilon(\omega)-1]/[\epsilon(\omega)+2]$, where $\epsilon(\omega)$ is the particles' permittivity. Assuming a Lorentz-Drude-like dispersion, $\epsilon(\omega) = 1 + \omega_p^2/(\omega_0^2 - \omega^2 - i\omega\Gamma_0)$ (being $\omega_p$ the plasma frequency, $\omega_0$ the natural frequency, and $\Gamma_0$ the damping constant), the polarizability can be written as [27,28]

$$\alpha(\omega) = \frac{4\pi a^3(\omega_F^2 - \omega_0^2)}{\omega_F^2 - \omega^2 - i\{\omega\Gamma_0 + 2(ka)^3(\omega_F^2 - \omega_0^2)/3\}}, \tag{3}$$

where $\omega_F$ is the Fröhlich resonance frequency given by $\omega_F^2 = \omega_p^2/3 + \omega_0^2$. The term $-i\Gamma_0\omega$ accounts for damping by absorption, whereas $-i2(ka)^3(\omega_F^2 - \omega_0^2)/3$ accounts for radiative damping [27] (see Refs. [29,30] for the subtle problem of radiative damping in two-level atoms).

*Interaction forces induced by a stationary random field.*—The particles are illuminated by an homogeneous and isotropic random light field consisting of a superposition of unpolarized and angularly uncorrelated plane waves (equivalent to $4\pi$ illumination with diffuse light [10]). In the presence of a quasimonochromatic stationary random field, the averaged force on particle "1" located at $\mathbf{r}$, due to the presence of particle "2" at the origin of coordinates, has been shown to be [9,10]:

$$\mathbf{F}_{12}(r) = \left\{\frac{4\pi U_E}{k^2}\right\}\sum_{i=x,y,z}\left[\text{Im}\left\{\frac{k^6\alpha^2 g_i g_i'}{1 - k^6\alpha^2 g_i^2}\right\} - \{k^2\sigma_{\text{abs}}\}\frac{\text{Re}\{k^3\alpha[g_i g_i' + g_i g_i'^*]\}}{|1 - (k^3\alpha)^2 g_i^2|^2}\right]\frac{\mathbf{r}}{r}, \tag{4a}$$

where $U_E = \epsilon_0\langle|\mathbf{E}(\mathbf{r},t)|^2\rangle/2$ is the time-averaged energy of the fluctuating electric field per unit of volume (this is half of the energy density of the electric *and* magnetic fields) and $g_i' = \partial g_i/\partial(kr)$ with

$$g_x(kr) = g_y(kr) = \frac{e^{ikr}}{4\pi kr}\left(1 + \frac{i}{kr} - \frac{1}{(kr)^2}\right) \tag{5}$$

$$g_z(kr) = \frac{e^{ikr}}{4\pi kr}\left(-\frac{2i}{kr} + \frac{2}{(kr)^2}\right). \tag{6}$$

It is worth to mention that the derivation of Eqs. (4) implicitly assumes that the system is in a nonequilibrium stationary state: the energy *absorbed* is transferred to a thermal bath and the contribution of the incoherent thermal emission to the force is assumed to be negligible.

*Forces between nonabsorbing particles.*—The results for nonabsorbing particles are summarized in Fig. 1 where we plot the force [normalized to $F_0 = U_E k^4|\alpha(\omega)|^2/(4\pi)$] versus separation distance for different illumination frequencies $kr = 2\pi r/\lambda \equiv R$. Forces were calculated from Eq. (4) using the polarizability given by (3). We compare the modulus of the actual force versus distance (in logarithmic scale) with the trends expected for $R^2$ and $R^{-2}$ in Fig. 1(a) (resonant case $\omega = \omega_F$) and $R^{-7}$ and $R^{-2}$ in Fig. 1(b) (off resonance, $\omega \ll \omega_F$). In order to understand these results, notice that Eq. (4) simplifies considerably in the weak scattering limit, $|(k^3\alpha)g_i|^2 \ll 1$, when recurrent scattering events, responsible of the denominators in Eqs. (4), do not play a relevant role. In absence of absorption, $\sigma_{\text{abs}} = 0$, the interaction force, Eq. (4a), exhibits an oscillatory behavior in the far-field zone ($kr \gg 1$) with an envelop that decays as $R^{-2}$:

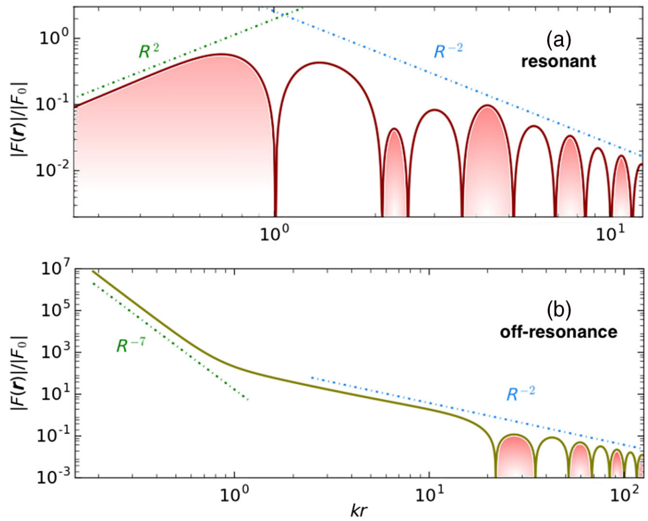


FIG. 1. *Forces between nonabsorbing particles*. Log-log plot of the absolute value of the interaction force between nonabsorbing particles with $\alpha$ given by Eq. (3), with $\omega_0 = 0.1\omega_F$, $\Gamma_0 = 0$, and $a = \lambda_F/100$ for (a) the resonant frequency $\omega = \omega_F$ and (b) strongly off resonance $\omega = 0.1\omega_F$. Red shadowed regions indicate repulsive interaction force.

$$\lim_{kr\gg 1} \mathbf{F}_{12}(r)|_{\text{No abs}} \sim -U_E \frac{k^4|\alpha|^2}{2\pi} \frac{\cos(2[kr + \delta_\omega])}{(kr)^2} \frac{\mathbf{r}}{r}, \quad (7)$$

a result that was first predicted [8] for the interactions between dipolar particles excited by a spatially coherent field after averaging over all orientations of the inter-atomic axis with respect to the incident beam. It is remarkable that these results, originally obtained using fourth order QED perturbation theory [8], are strictly equivalent to those obtained from a classical electrodynamic approach [9,10].

Strongly off resonance ($\omega \ll \omega_F$), the weak scattering approximation holds even at near field distances ($kr \ll 1$) since $|(k^3\alpha)g_i|^2 \sim (a/r)^6 \ll 1$, as long as $a \ll r$, and

$$\lim_{ka\ll kr\ll 1} \mathbf{F}_{12}(r,\omega)|_{\text{No abs}} \sim -U_E \frac{k^4|\alpha|^2}{4\pi} \left\{ \frac{22}{15} \frac{\cos(2\delta_\omega)}{(kr)^2} + 18 \frac{\sin(2\delta_\omega)}{(kr)^7} \right\} \frac{\mathbf{r}}{r}. \quad (8)$$

Previous works [8,9,23] disregard the last term assuming that, far from resonance, the imaginary part of the polarizability can be neglected (i.e., $\sin 2\delta_\omega \sim 2\delta_\omega \sim 0$ and $\cos 2\delta_\omega \sim 1$), which would lead to an attractive $r^{-2}$, gravity-like, interaction force at short distances. However, in absence of absorption we have $\sin\delta_\omega = k^3|\alpha|/(6\pi) \sim 2(ka)^3/3$, i.e., $\sin 2\delta_\omega \sim 4(ka)^3/3$. This implies that, for small distances, the attractive term $\sim r^{-7}$ dominates the interaction. This explains the crossover from $R^{-2}$ to a $R^{-7}$ tendency observed in Fig. 1(b).

The interaction force in the near field is strikingly different at the resonant condition $\omega = \omega_F$. In absence of absorption $\alpha(\omega_F) = i6\pi k_F^{-3}$ and, in the near field, the interaction force is dominated by recurrent scattering contributions since $|(k^3\alpha)g_i|^2 \sim |6\pi g_i|^2 \gg 1$. In this limit the near-field force is repulsive and proportional to the energy density of the random field,

$$\lim_{kr\ll 1} \mathbf{F}_{12}(r,\omega_F)|_{\text{No abs}} = 12\pi U_E r^2 \frac{\mathbf{r}}{r}, \quad (9)$$

being independent on the actual resonant frequency, $\omega_F$, or any other particle's property. This universal limit had not been noticed previously. For dielectric nanoparticles this would be valid for distances as small as $r \sim 3a$ (for $r < 3a$ high order multipoles start being relevant).

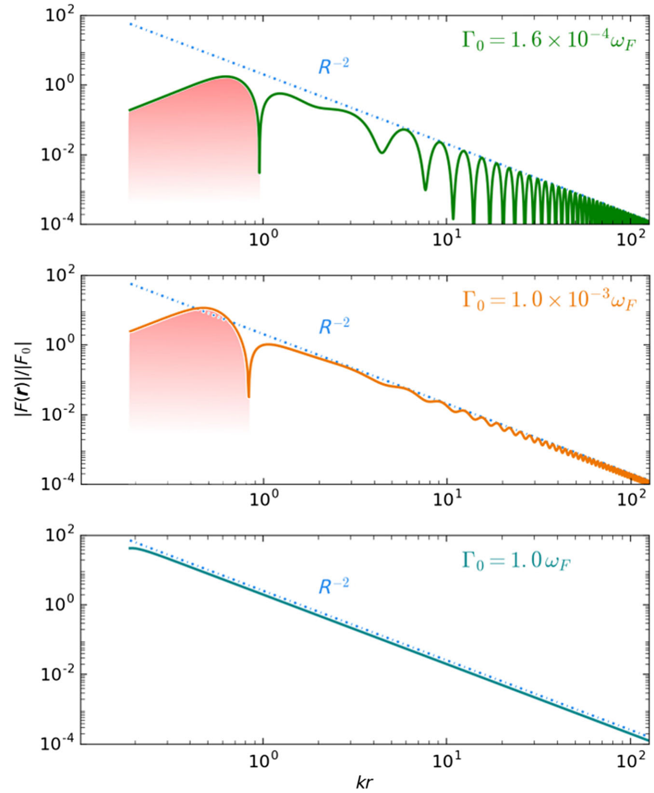


FIG. 2. *Forces between absorbing particles*. Log-log plot of the absolute value of the interaction force between absorbing particles with $\alpha$ given by Eq. (3), with $\omega_0 = 0.1\omega_F$, $\omega = \omega_F$, and $a = \lambda_F/100$ for different damping coefficients (shown in each plot). Red shadowed regions indicate repulsive interaction force.

*Forces between absorbing particles.*—The interaction forces at resonance are strongly modified by the presence of absorption. Figure 2 summarizes the behavior of the interaction forces at the resonant condition $\omega = \omega_F$ for different values of the damping coefficient $\Gamma_0/\omega_F$. As it can be seen, the oscillatory behavior of the force disappears as absorption increases and, eventually, the interaction turns gravitational-like at all separation distances all the way from the near to the far field zones!

A simple analytical expression for the force can be obtained in the limit when the extinction cross section is dominated by absorption [26], i.e., $\sigma_{\rm abs} \sim \sigma_{\rm ext}$. In the weak scattering limit the force, can be written as

$$\mathbf{F}_{12}(r)|_{\rm abs} \sim 4\pi U_E k^4 |\alpha|^2 \sum_{i=x,y,z} \left[ \mathrm{Im}\left\{ \frac{e^{2i\delta_\omega}+1}{2} g_i g_i' \right\} - \mathrm{Im}\left\{ \frac{e^{2i\delta_\omega}-1}{2} g_i g_i'^* \right\} \right] \frac{\mathbf{r}}{r}, \quad (10)$$

where the first term, proportional to the real part of the polarizability, is the so-called gradient force (proportional to the gradient of the exciting field intensity on each particle) and presents an oscillatory behavior reminiscent of the Fabry-Perot-like interference. The second term, proportional to the imaginary part of the polarizability, corresponds to the "radiation pressure" force [31] (proportional to the incoming linear momentum flux on each particle), which, in our case, is dominated by absorption. This last term can be seen as the shadow effect reminiscent of Fatio de Duillier–LeSage corpuscular theory (notice that, in the far-field, the $g_i g_i'^*$ factor in the absorption term does not oscillate). At the Fröhlich resonance, $\delta_{\omega_F} = \pi/2$, the gradient force vanishes and the interaction force becomes

$$\mathbf{F}_{12}(r, \omega_F)|_{\rm abs} \sim -U_E \frac{k_F^4 |\alpha(\omega_F)|^2}{2\pi} \frac{1}{(k_F r)^2} \frac{\mathbf{r}}{r}; \quad (11)$$

i.e., a force that is a nonoscillating long range gravitylike interaction. Notice that, at the Fröhlich resonance, the weak scattering limit implies

$$\left(\frac{a}{r}\right)^6 \left(\frac{\omega_F^2 - \omega_0^2}{\Gamma_0 \omega_F}\right)^2 = \left(\frac{a}{r}\right)^6 Q^2 \ll 1. \quad (12)$$

Equation (11) will then hold for distances as small as $r \sim 3a$ as long as the quality factor, $Q$, of the resonance remains smaller than $\sim 30$.

In order to check the validity of the results in a realistic scenario, we consider silver nanoparticles with a nearly ideal free electron Lorentz-Drude-like dielectric response. Similar results would apply to alkali metals or aluminum plasmon nanoparticles or to small particles presenting surface plasmon-polariton modes [like silicon carbide (SiC) and some other insulating solids with $\lambda_F \sim 10\ \mu$m [26]], as well as to more complex hybrid systems tuned to support Fröhlich resonances. In Fig. 3 (top panel) we plot the polarizability of 5 nm sized Ag plasmon particles, computed from the first Mie coefficient using experimental values of $\epsilon(\omega)$ [32], which is well described by Eq. (3) [33]. Note how the real part of the polarizability is equal to zero, i.e., the phase shift $\delta_\omega = \pi/2$, at 317 nm and 352 nm (in contrast, the real part of the polarizability of gold nanoparticles does not vanish [33]). Gravitylike interactions at these two wavelengths are indeed observed as shown in Fig. 3 where we plot the force given by Eq. (4) at different wavelengths. For larger sizes, up to approximately 20 nm radius the optical response is still dipolar and highly

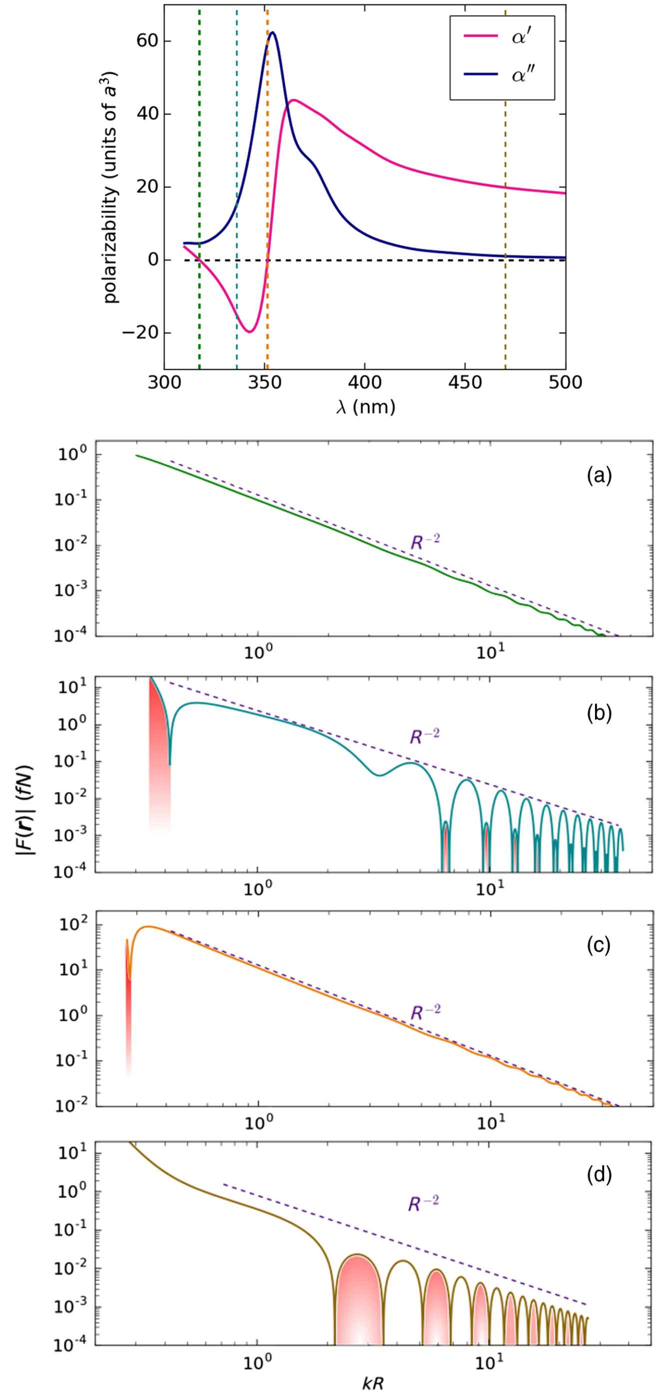


FIG. 3. *Forces between silver nanoparticles*. (top) Real (black line) and imaginary part (red line) of the polarizability versus wavelength in vacuum for a silver nanoparticle with $a = 5$ nm. Log-log plots of the force between two silver nanoparticles calculated for a power density of 10 W/$\mu$m$^2$ at different wavelengths (vertical dashed lines): (a) $\lambda = 317$ nm, (b) $\lambda = 337$ nm, (c) $\lambda = 352$ nm, and (d) $\lambda = 47$ nm. Red shadowed regions indicate repulsive forces.

absorbing but gradually departs from the ideal behavior of Eq. (3).

It is interesting to compare the relative magnitude of Mock gravity forces, $F_{\mathrm{MG}}$, with other relevant forces like van der Waals (vdW) forces. For a Hamaker constant, $A \sim 4 \times 10^{-19}$ J, typical for metals, the vdW interaction force at a separation distance $r \sim 6a = 30$ nm is given by [11] $F_{\mathrm{vdW}} \sim -(32/3)Aa^6/r^7 \sim -4 \times 10^{-5} A/a$ which, for $a = 5$ nm gives $\sim 3$ fN forces. At the same distance and $\lambda \sim 352$ nm [Fig. 3(c)], these forces are of the same order of magnitude as $F_{\mathrm{MG}}$ for power densities of the order of 1 W/$\mu$m$^2$. However, the force (power density) can be dramatically increased (reduced) by considering larger particles. For $a = 20$ nm and $r \sim 6a = 120$ nm, $F_{\mathrm{vdW}} \sim 1$ fN we obtain similar $F_{\mathrm{MG}}$ at 1 mW/$\mu$m$^2$ while, for 1 W/$\mu$m$^2$, Mock gravity forces would reach values of the order of pN. In a liquid environment, electrostatic double layer forces, coming from the interaction with the ions of the solvent, could also compete with optical forces [34] but can be cancelled by setting an isoelectric point at which the so-called zeta potential is zero (see e.g., [35] for Ag nanopartiples in aqueous environments). Moreover, Fröhlich resonances can be significantly enhanced and shifted when the particles are immersed in lossless solvents [26].

Unlike actual gravitational forces, light induced forces are generally not pairwise additive. However multiple scattering effects can be strongly reduced at the Fröhlich resonance, when the particles extinction cross section is dominated by absorption, and we then expect pairwise interactions to be a good approximation even for relatively high particle densities, $\rho$ (see the Supplemental Material at [36]). This can be relevant in colloidal suspensions since, even if the forces between two particles can be small at low power densities, the actual force on each single particle involves all the particles in the system. This holds as long as the typical absorption length, $\sim 1/(\rho\sigma_{\mathrm{abs}})$, is larger than the size of the cloud of particles. Otherwise light would be mainly absorbed by the external layers inducing nontrivial light intensity (and temperature) gradients whose theoretical analysis is well beyond the scope of this Letter.

*Summary.*—We have analyzed the interaction forces between identical resonant nanoparticles illuminated by a quasimonochromatic isotropic random light field. We found two main, unexpected, force regimes. First, in absence of absorption, when the light frequency is tuned to the particles' resonance the near-field force ($kr \ll 1$) is repulsive and increases with separation as $r^2$. Interestingly, the near-field force at this resonance is proportional to the energy density of the random field, but it does not depend on the actual frequency or any other particle's property. The second regime is relevant for highly absorbing particles. When the frequency of the random field is tuned to the particles' Fröhlich resonance, we found that the interaction law follows an ideal, nonoscillating, $\sim 1/r^2$ law all the way from near to far-field separation distances. This is it is in contrast with the oscillatory behavior predicted for lossless resonant particles by all previous works based on both QED and classical approaches. Our derivation assumes that light-particle interactions take place in free space or homogeneous media. However, the radiative coupling can be strongly modified in structured environments, as was early shown for atoms [37], or in confined geometries [38]. We believe that our results will stimulate further work to explore the nature of random-light-induced interactions in structured media.

Our work suggest novel approaches in the study of (mock) gravitational interactions opening a promising laboratory for testing the intriguing predictions of the statistical mechanics of systems with long-range interactions [39,40].

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